Atmos. Chem. Phys. Discuss., 8, S8947–S8952, 2008 www.atmos-chem-phys-discuss.net/8/S8947/2008/ © Author(s) 2008. This work is distributed under the Creative Commons Attribute 3.0 License.



ACPD 8, S8947–S8952, 2008

> Interactive Comment

Interactive comment on "Distribution of VOCs between air and snow at the Jungfraujoch high alpine research station, Switzerland, during CLACE 5 (winter 2006)" *by* E. Starokozhev et al.

E. Starokozhev et al.

Received and published: 11 November 2008

Comment: One of my major concerns is related to the days March 11, 12 that were declared as initial state of the thermodynamic system atmosphere. The true initial state conditions prior to the snowfalls on Feb 24 and Mar 01 may have been different to the conditions on Mar 11, 12. The Jungfraujoch presumably receives a large fraction of its atmospheric pollution from nearby industrialized urban regions. The wind conditions and oxidative capacity of the atmosphere may have been different and accordingly the atmospheric VOC concentrations as well. Another question is related to the use of fluxes to calculate the removal efficiencies instead of using just atmospheric gaseous concentrations. The reasoning why the amounts of organic chemicals in the gas phase can be better described using their fluxes, is not described in a conclusive way. In-



cidentally, it is not mentioned where the wind speed was measured, at which height above the ground and in which distance to the other measurements. Especially in high alpine regions wind conditions are very heterogeneous.

Response: The atmosphere as a complex thermodynamic system can be described by means of lots of parameters, which can vary extremely. We have chosen the group of parameters, that characterize the conditions of physical chemical process (transformation) of substances in the atmosphere, which were identical in the chosen days, that gave us an opportunity to compare the other parameters (for ex. chemical fluxes). The Reviewer has noticed absolutely correct, the wind conditions can be different and influence the VOC concentrations in the atmosphere. Because of it we suggest to describe the amounts of VOC in the atmosphere by means of the more complex parameter, that includes the quantitative characteristic of the chemicals in system (concentration) as well as the parameter of system (wind speed), that determines transport velocity of chemicals. The meteorological information was obtained from the Swiss Meteorological Institute. The meteorological parameters were measured with the help of automatically whether station of MeteoSwiss, which was installed on the Sphinx's platform as well as the inlet of AirmoVOC

Comment: (14348, 6) It should be clearly stated which fluxes are referred to. Response: It referred to the fluxes of compounds in the atmosphere, which concentrations we've measured by AirmoVOC in ambient air.

Comment: (14348, 10) efficiency of removal from the atmosphere should clearly be mentioned. Response: It will be improved by us to: The fluxes were used to calculate the efficiency of removal of VOCs by snow, which we defined as difference between the initial and final flux values of compounds before and after wet deposition.

Comment: (14349, 17 - 20) What information does this sentence provide? (14349, 20 - 23) This sentence should be a little more scientifically accurate. (14349, 25 - 28) Research has been earlier conducted (e.g. Schöndorf and Herrmann, 1987). (The

Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



references Hoff et al., 1995 and Wania et al., 1999 describe experimental and modeling work and are not suitable to substantiate the detection of organic pollutants in remote areas.

Response: The sentence (14349, 17 -20) will be deleted. The citation (14349, 20 - 23) will be included in the paper in its original version. At high latitudes and altitudes, such deposition of VOCs is greatly influenced by low temperatures and in particular the phase transition of water at temperatures below 0°C (Lei and Wania, 2004) The sentences (14349, 25 - 28) refer to the Roth's paper and will be improved in the final paper. Research has been conducted aimed at understanding the processes linked to the occurrence of organic pollutants in snow and ice since organic pollutants were detected in snow in remote areas and at a high alpine site (Roth et al., 2004, Gröllert and Puxbaum, 2000).

Comment: (14350, 9) Aerosols are contaminants?

Response: 'Aerosol' will be improved to 'Aerosol particles'

Comment: (14350, 18- 19) this is already included in the statement made two sentences before.

Response: This one will be deleted

Comment: (14351, 12 - 14) the chemical flux towards higher altitudes in the atmosphere can also be influenced by strong upward fluxes of air masses. (14351, 23 - 25) (Eq.3) the influence of chemical partitioning between the phases is missing in this equation.

Response: Of course, the vertical transport is influenced by lots of other factors, because of it we've used the proportionality sign. By means of this equation we wanted to show that the main process of the vertical transport of chemicals is diffusion, determined by the gradient of concentration between the source and accepter. The influence of chemical partitioning between the phases is indirectly described by density of 8, S8947–S8952, 2008

Interactive Comment



Printer-friendly Version

Interactive Discussion



precipitation, which is proportional to compounds concentration in precipitation.

Comment: (14352, 10 - 13) this coefficient will never approach infinity. Better use the expressions high values and/or low values.

Response: To determine the boundary conditions are used the statements: approach infinity for high values and approach 0 for low values, because of it we used those statements to show the boundary conditions of partitioning coefficient.

Comment: (14353, 10) is the location of the inlet uninfluenced by possible emissions from the building?

Response: Inlet was installed on the Sphinx's platform 5 m above the bottom to exclude the influence by possible emissions from the building.

Comment: (14356, 4) Here and several times later in the text the authors refer to water as the condensed phase in the system air-snow. If the authors refer to the quasi-liquid layer at the surfaces of the snow grains, the properties of water and the quasi-liquid layer are too different to let water act as a substitute for the QLL.

Response: Here is meant the water which forms its own phase in the atmosphere. In the other parts of the paper we also mentioned the adsorption layer of condensed water at the surface of the snow.

Comment: (14357, 6) with decreasing temperature (14357, 6) What does it mean: enhances air/ice partitioning? This sentence should be written in a more accurate way. (14357,7) low temperatures -This is already said within the previous sentence

Response: It will be improved in the following way: According to Lei & Wania (2004), one possible explanation for this observation might be cold temperature removal of organic chemicals due to a reduction in vapor pressure with temperature, which can influence the deposition mechanism and consequently air/ice partitioning".

Comment: (14359, 2 - 6) this is not really an explanation. (14359, 22 - 24) For com-

8, S8947–S8952, 2008

Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



pounds with a low distribution coefficient (Kair/snow), sorption to the snow surface should play a dominant role.

Response: We suggest this explanation as one of the variants and it will be extended. On one hand calculated coefficients can differ from the empirical coefficients, because a specific snow surface area in equation (6) may largely differ from that of the snowflakes during snowfall. On the other hand, according to Roth (2004) that is, however, the evidence that experimental sorption data cannot always be described solely by physical adsorption. It also indicates that a combination of processes like dissolution in the bulk ice or in a surface layer, incorporation of gases into the crystal lattice during crystal growth, particle scavenging (nucleation and impaction scavenging by growing ice crystals) (Fries, 2008) might contribute to the overall sorption on ice. For the theoretical understanding of the various sorption processes in and on snow, additional laboratory investigations should be conducted. Comment: (14359, 22 - 24) will be written in more accurate way.

Response: For compounds with low water solubility and a low distribution coefficient, removal from the air by snow is not to be described only by physical adsorption.

Comment: Table 1: Why are the meteorological parameters listed for March 03 but not for March 12. The latter date is important because it represents the initial state chosen by the authors.

Response: It's a technical mistake that will be improved.

Comment: (14364) Table 2: Rather use the SI unit for the Henrys law constant.

Response: We will use the SI unit (Pa.m3/mol) for the Henrys law constant

Comment: (14365) Table 3: The calculations used in Lei and Wania (2004) assume a specific snow surface area which may largely differ from that of the snowflakes during snowfall in this field study. Specific snow surface areas are highly variable. Response: The Reviewer is absolutely right specific snow surface areas are highly variable, we

8, S8947–S8952, 2008

Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



have chosen the value for fresh snow (1,0 m2/g) that we have found in the literature (Franz and Eisenreich, 1998)

Comment: (14368) Fig. 3: Maybe it is more meaningful to show the chemicals at the x-axes of both plots in the same order.

Response: The labelling of the substances (x-axis) in Fig. 3 can not be identical because the initial flux values are shown in decreasing order and do not show the same pattern.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 14347, 2008.

ACPD

8, S8947–S8952, 2008

Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

